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Hydrogen Evolution **Hot Paper**

Silver Single Atom in Carbon Nitride Catalyst for Highly Efficient Photocatalytic Hydrogen Evolution

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Abstract: *Single atom catalysts (SACs) with the maximized metal atom efficiency have sparked great attention. However, it is challenging to obtain SACs with high metal loading, high catalytic activity, and good stability. Herein, we demonstrate a new strategy to develop a highly active and stable Ag single atom in carbon nitride (Ag-N_2C_2/CN) catalyst with a unique coordination. The Ag atomic dispersion and Ag-N_2C_2 configuration have been identified by aberration-correction high-angle-annular-dark-field scanning transmission electron microscopy (AC-HAADF-STEM) and extended X-ray absorption. Experiments and DFT calculations further verify that Ag-N_2C_2 can reduce the H_2 evolution barrier, expand the light absorption range, and improve the charge transfer of CN. As a result, the Ag-N_2C_2/CN catalyst exhibits much better H_2 evolution activity than the N-coordinated Ag single atom in CN (Ag-N_4/CN), and is even superior to the Pt nanoparticle-loaded CN (Pt_{NP}/CN). This work provides a new idea for the design and synthesis of SACs with novel configurations and excellent catalytic activity and durability.*

Single atom catalysts (SACs), which combine the advantages of homogeneous and heterogeneous catalysts, have attracted a great deal of interest.[1] To date, functional SACs have been explored for application in various fields such as electrocatalysis,[2] industrial catalysis,[3] and photocatalysis,[4] owing to their maximized metal atom utilization, high selectivity, and low reaction energy barriers.[5] However, SACs suffer from low metal loading and poor stability, making them inefficient and unsustainable. To improve their catalytic efficiency and stability, some strategies have been developed, such as in situ anchor strategy[6] and heteroatom or rich defect strategies.[7] However, these approaches still cannot resolve the abovementioned problems because of the limited heteroatom and defect content.[8] Therefore, new approaches must be developed to obtain SACs with enhanced catalytic activity and stability.

Carbon nitride (CN) is expected to be a promising metal single atom support owing to its good coordination ability and ultrahigh content of N atoms. Notably, the lone electron pairs on N make it easier for the metal atoms to coordinate with N than C, resulting in the coordination of most metal atoms of SACs with N to form M-N_x (M: metal atom).[9] Furthermore, the excited electrons of CN are mainly distributed in the C atoms because the C atoms chiefly contribute to the bottom of conduction band (CB).[10] As a result, the excited electrons of CN in the CB need to overcome a high energy barrier to transfer to metal atoms when the SACs have M-N_x coordination. Especially, if SACs are fabricated using metals with low work functions in CN, it becomes difficult for the SAC to transfer its excited electrons from CN to metal atoms with M-N_x coordination, leading to poor catalytic activity.[11] Therefore, it is imperative to regulate the C and N co-coordinated metal single atom to reduce the transfer barrier for excited electrons from CN to metal single atom, thereby realizing evidently improved catalytic activity.

Herein, consideration the low work function of Ag, we propose a supermolecular method to synthesize a Ag-N_2C_2/CN catalyst with a new M-N_2C_2 configuration, showing high metal loading amount, excellent photocatalytic activity, and good stability (Figure 1 a). Experimental and DFT calculation results showed that C and N co-coordinated Ag could reduce the H_2 evolution overpotential, expand the visible light absorption range, and promote charge separation and migration of CN. These multiple effects of the Ag single atom resulted in the Ag-N_2C_2/CN with efficient photocatalytic H_2 evolution and remarkable stability, superior to those of Pt_{NP}/CN. The strategy of using C and N co-coordinated metal single atom provides a new idea to simultaneously enhance the metal loading and dispersibility as well as the catalytic activity and stability of SACs.

The supermolecule formed by self-assembly via hydrogen bonds (Figure S1, Supporting Information) can uniformly distribute the Ag atoms (Figure S2), increase the melting point of the raw materials (Figure S3), control the content of single atom Ag (Figure S4), and increase the surface area of CN (Figure S5), which are crucial for better dispersibility and higher loading of Ag atoms in CN. The introduction of single atom Ag did not affect the structure of CN (Figure S6 and S7). In Ag-N_2C_2/CN, no Ag nanoparticles (AgNPs) can be

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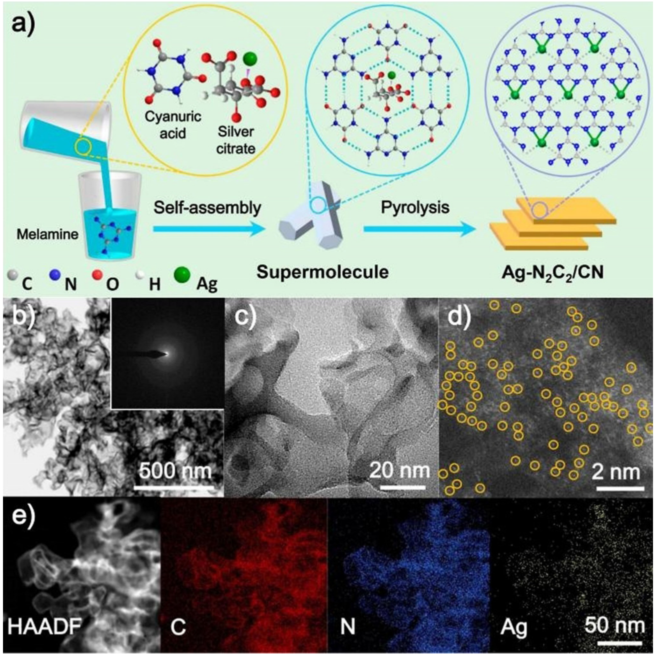

Figure 1. a) Schematic illustration of preparation of Ag-N_2C_2/CN, b) TEM (inset SAED), c) HR-TEM, d) AC-HAADF-STEM (the bright dots in the yellow circles are Ag single atoms), and e) HAADF-STEM with elemental mapping images of Ag-N_2C_2/CN.

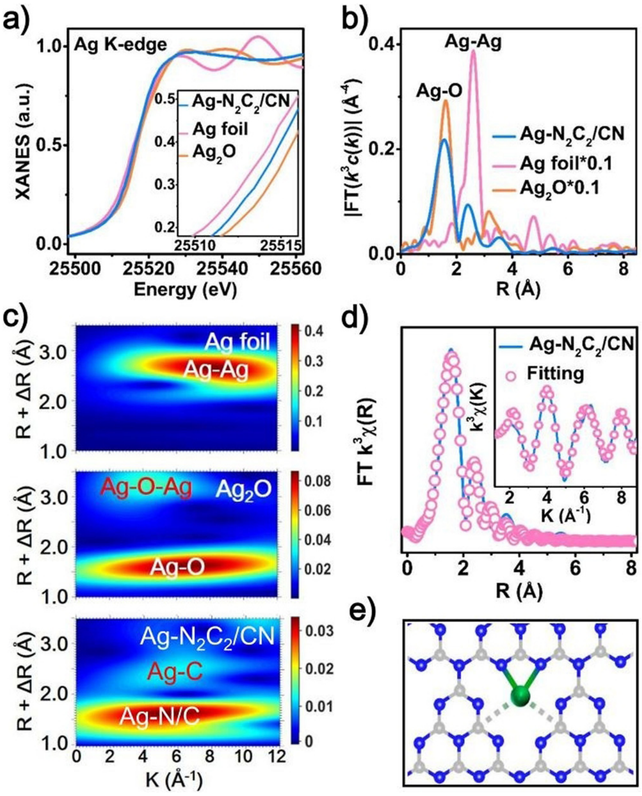

Figure 2. a) Ag K-edge XANES, b) Fourier transform of Ag K-edge EXAFS spectra, c) WT of Ag foil, Ag_2O, and Ag-N_2C_2/CN. d) EXAFS fitting curve in R space and e) structure model of Ag-N_2C_2/CN (Ag green, N blue, C grey).

observed in the high-resolution transmission electron microscopy (HR-TEM) image (Figure 1 b,c). The selected area electron diffraction (SAED) can also confirm that there is no crystalline Ag (inset in Figure 1 b) in Ag-N_2C_2/CN, in line with the XRD analysis (Figure S6). The AC-HAADF-STEM image of Ag-N_2C_2/CN shows bright dots (Figure 1 d), which are identified as the atomically dispersed Ag in CN. The uniform dispersion of Ag atoms in CN was verified by elemental mapping (Figure 1 e). In addition, the vertical element analysis of Ag-N_2C_2/CN demonstrates that Ag atoms were also uniformly distributed in CN (Figure S8). As for Ag-N_4/CN, the atomically dispersed Ag was also confirmed by elemental mapping and AC-HAADF-STEM images (Figure S9).

To further confirm the Ag atoms dispersion and coordination environment, X-ray absorption near edge structure (XANES) and extended X-ray absorption fine structure (EXAFS) were carried out. For the Ag K-edge spectra, the absorption threshold of Ag-N_2C_2/CN between Ag foil and Ag_2O (Figure 2 a) demonstrates the oxidation state of Ag ($Ag^{\delta+}$, $0 < \delta < 1$) in Ag-N_2C_2/CN because of the strong interaction between the single atom Ag and CN, consistent with the X-ray photoelectron spectroscopy results (Figure S10). The Fourier-transformed k_3-weighted EXAFS (Figure 2 b) image shows two main peaks at 1.55 and 2.41 Å, corresponding to the first coordination shell of Ag-N and the second coordination shell of Ag-C, respectively. Compared with the Ag foil and Ag_2O, no Ag-Ag and Ag-O coordination peaks at 2.59 and 1.62 Å were observed, demonstrating that the atomically dispersed Ag was coordinated with both C and N in CN. Furthermore, only one intensity maximum at 5.7 $Å^{-1}$ can be observed from the wavelet transform (WT) contour plots of Ag-N_2C_2/CN (Figure 2 c), which can be assigned to Ag-N/C coordination without Ag-Ag signal. According to the EXAFS fitting results, the coordination numbers of Ag-N and Ag-C are 2.3 and 1.8 (Figure 2 d and Table S1), respectively. These results demonstrate that the Ag single atom in CN adopts the Ag-N_2C_2 configuration (Figure 2 e). Therefore, it can be concluded that the novel coordinated Ag-N_2C_2/CN was successfully synthesized by the supermolecular method.

As depicted in Figure 3 a, AgNPs do not affect the light absorption of CN; however, the Ag single atoms of Ag-N_2C_2 and Ag-N_4 can expand the light absorption range and reduce the band gap of CN (inset in Figure 3 a). In addition, the strongest photoluminescence (PL) quenching, shortest PL lifetime, and smallest electrochemical impedance spectrum arc radius of Ag-N_2C_2/CN (Figure 3 b–d) prove that Ag-N_2C_2/CN has the least photogenerated charge recombination, fastest charge transfer, and minimum charge transfer resistance among the as-prepared catalysts. Moreover, the highest photocurrent of Ag-N_2C_2/CN can provide the most photogenerated charge for the redox reaction (Figure 3 e). As shown in Figure 3 f, the loading of Ag can reduce the H_2 evolution overpotential, where Ag-N_2C_2/CN has a smaller initial voltage and a higher current density than others. These results indicate that Ag-N_2C_2 has the lower charge transfer barrier than that achieved with AgNPs and Ag-N_4.

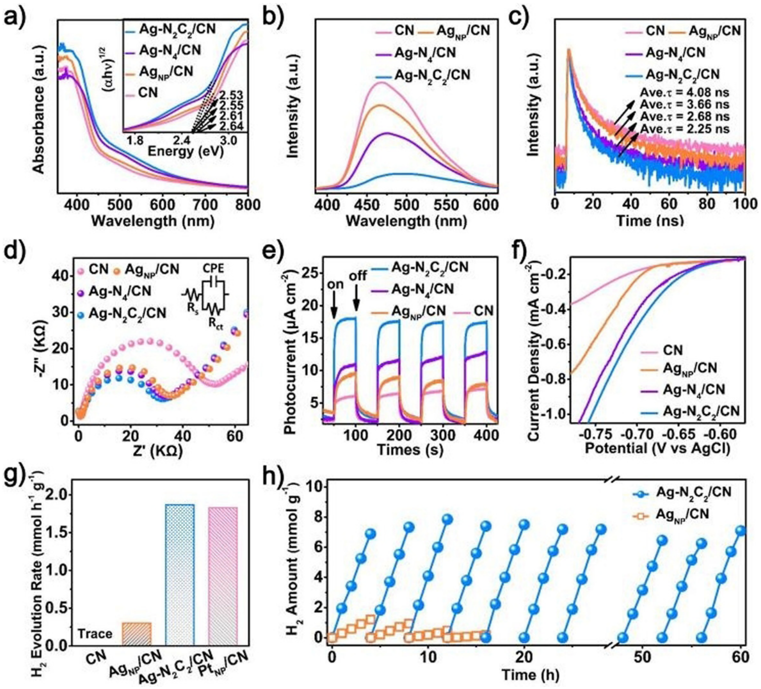

Figure 3. a) UV-vis absorption spectra and Tauc plots (inset), b) PL spectra, c) time resolution PL spectra, d) electrochemical impedance spectra, e) photocurrent response, and f) linear sweep voltammetry plots of CN, Ag$_{NP}$/CN, AgN$_4$/CN, and Ag-N$_2$C$_2$/CN; g) H_2 generation rates of as-prepared samples, and h) the cycling H_2 evolution activity of the Ag$_{NP}$/CN and Ag-N$_2$C$_2$/CN.

Next, the photocatalytic H_2 evolution performance of the samples was investigated. The co-catalysis effect of AgNPs remarkably enhanced the photocatalytic H_2 evolution rate of CN (Figure 3 g).[12] When AgNPs were replaced by Ag-N$_4$, the photocatalytic H_2 evolution rate slightly increased (Figure S11a). Ag-N$_2$C$_2$/CN shows a much higher photocatalytic H_2 evolution rate than Ag$_{NP}$/CN and Ag-N$_4$/CN; in fact, it is even better than that achieved with Pt$_{NP}$/CN (Figure 3g and S11b). Compared with the literature on Ag-loaded photocatalysts, Ag-N$_2$C$_2$/CN has a much higher photocatalytic H_2 evolution activity (Table S2). Moreover, the Ag-N$_2$C$_2$/CN almost maintains its photocatalytic H_2 evolution activity at the initial level even after 15 cycles (Figure 3 h). In contrast, Ag$_{NP}$/CN nearly loses its photocatalytic activity after four cycles only. Notably, the structure and chemical state of Ag-N$_2$C$_2$/CN did not change before and after the cycling test (Figure S12). These results prove the excellent photocatalytic activity and stability of Ag-N$_2$C$_2$/CN.

DFT calculations were employed to further understand the excellent performance of Ag-N$_2$C$_2$/CN. The Ag single atom can expand the light absorption range of CN by reducing the band gap rather than introducing an impurity level (Figure S13). According to the density of state (DOS), the CB of Ag-N$_2$C$_2$/CN is closer to the Fermi level than that of CN (Figure 4 a), indicating that Ag-N$_2$C$_2$/CN displays a higher carrier density and better charge transfer capability.[13] The differential charge density diagrams show that Ag-N$_2$C$_2$/CN is more conducive to charge distribution than Ag$_{NP}$/CN and Ag-N$_4$/CN (Figure 4 b), leading to a faster electron transfer from CN to Ag.[14] Figure 4 c shows that in an alkaline environment, Ag-N$_2$C$_2$/CN has the lowest energy barrier from H_2O to HO-H, and then to H*, indicating that Ag-N$_2$C$_2$/CN has the best H_2 evolution process among the as-prepared catalysts (Figure S14).[15] The photocatalytic enhancement mechanism of Ag-N$_2$C$_2$/CN is proposed based on experimental tests and DFT calculations (Figure 4 d). First, the smaller band gap of Ag-N$_2$C$_2$/CN expands the light absorption range, which can increase the amount of photo-induced electrons in CB. Second, the C and N co-coordination can reduce the electron transfer barrier from CN to Ag single atom. Finally, the reduced H_2 evolution overpotential from the Ag single atom can accelerate the reaction rate of H^+ reduction. These results indicate that Ag-N$_2$C$_2$/CN shows excellent photocatalytic H_2 evolution performance.

In summary, a simple supermolecular strategy is developed to prepare Ag-N$_2$C$_2$/CN photocatalysts with novel configurations, high Ag loading, high activity, and high stability. Experimental characterizations and DFT calculations showed that the single atom dispersion and C and N co-coordination of Ag boost the photocatalytic H_2 evolution of CN by expanding the visible light absorption range, regulating the electronic structure, reducing the H_2 evolution overpotential, and improving the charge transfer. Impressively, the photocatalytic H_2 evolution activity of Ag-N$_2$C$_2$/CN was

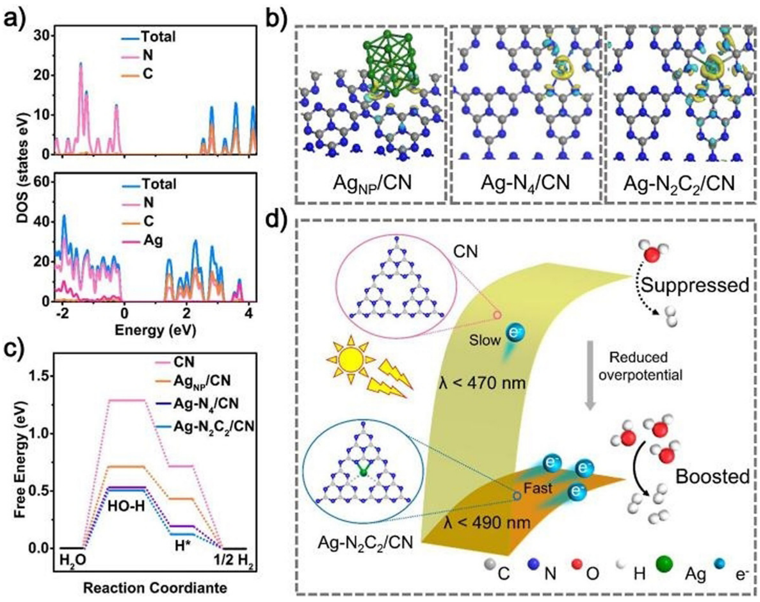

Figure 4. a) DOS of CN (top) and Ag-N$_2$C$_2$/CN (bottom), b) charge density difference maps, c) free energy profiles for H_2 evolution reactions over the as-prepared catalysts, and d) proposed mechanism for photocatalytic H_2 evolution over CN and Ag-N$_2$C$_2$/CN.

significantly higher than that of $\mathrm{Ag_{NP}}$/CN, and even better than that of $\mathrm{Pt_{NP}}$/CN. Besides, Ag-$\mathrm{N_2C_2}$/CN possesses much better photocatalytic stability than $\mathrm{Ag_{NP}}$/CN. This work provides a new idea for the design and synthesis of SACs with high metal loading, excellent catalytic activity, and high durability by adjusting the coordination modes of single atom metals.

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Conflict of interest

The authors declare no conflict of interest.

Keywords: co-coordination · electron transfer · photocatalysis · single-atom catalyst

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